

### **REMARKS**

#### **A. Status of the Claims and Claims Amendments**

In the Office Action of March 12, 2010, the status of the claims was as follows:

(1) Claims 65 and 77 were rejected under 35 U.S.C. §112, second paragraph, as being indefinite.

(2) Claims 64-69, 73-81 and 85-89 were rejected under 35 U.S.C. §102(e) as being anticipated by U.S. Pat. No. 6,565,231 (Cok '231).

(3) Claims 64-67, 69, 71-79, 81 and 83-90 were rejected under 35 U.S.C. §102(b) as being anticipated by Japanese Pat. Publ. JP 2001-043976 (Fukuyama '976).

(4) Claims 70-72, 82-84 and 90 were rejected under 35 U.S.C. §103(a) as being unpatentable over the Cok '231 reference.

(5) Claims 68, 70, 80 and 82 were rejected under 35 U.S.C. §103(a) as being unpatentable over the Fukuyama '976 reference in view of the Kido '209 patent.

Various amendments and additions to the claims have been made in this Amendment and Response to address the Examiner's rejections and/or to better distinguish this invention over the cited prior art. As will be appreciated in the following summary of the claim amendments, no new matter has been added. All of the claim amendments and new claims are fully supported by the original disclosure.

In this Amendment and Response, Claims 65 and 77 (and also Claim 88) have been amended to overcome the Sec. 112 rejection. Independent Claim 64 has been amended to incorporate the recitation of dopants from Claim 65. Similarly, independent Claim 75 has been amended to incorporate the recitation of dopants from Claim 77.

New apparatus Claim 91 has been added. New apparatus Claim 91 is based on Claim 64, but it is directed to the specific invention embodiment wherein the electroluminescent layer of the device consists of zirconium quinolate ( $ZrQ_4$ ) with diphenylquinacridine (DPQA) as the dopant in the claimed proportions. This specific invention embodiment is taught by Examples 1, 3, 4, 5, 8, 9 and 10 of the Specification, and it is the invention embodiment on which Figs. 37/49 to 47/49 of the application are based. These Figures present comparative graphs that clearly demonstrate the performance superiority of EL devices fabricated according to Claim 91 when compared with comparable EL devices fabricated according to prior art techniques using aluminum quinolate.

New dependent Claim 92, dependent on Claim 91, recites the specific device structure recited in Example 1. Claims 78-86 have been made to depend, directly or indirectly, on new Claim 91. Claims 77 and 88 have been amended to recite that the dopant is diphenylquinacridine (DPQA); and, Claims 65 and 76 have been amended to recite that the metal quinolate is hafnium quinolate.

Applicants respectfully request that the Examiner reconsider and withdraw the various claim rejections based on the amendments to the claims and on the following Remarks.

**B. Sec. 102(e) Rejection – Cok ‘231**

Claims 64-69, 73-81 and 85-89 were rejected under 35 U.S.C. §102(e) as being anticipated by the Cok ‘231 patent (para. 10 of the Office Action of March 12, 2010). Reconsideration of this rejection is respectfully requested for the following reasons:

1. Aluminum Quinolate Was the Preferred Host Material According to Prior Art Teachings.

The classic technical paper on the use of doped aluminum quinolate is that of Tang et al, *J. Applied Physics*, Vol. 65, No. 9, 1 May 1989, 3910-3916 (a copy of which accompanied the previously filed Supplemental IDS). The reasons for one to select aluminum quinolate as the host material for electroluminescent applications are disclosed in the Tang paper at page 3911, right hand column, namely that: (1) aluminum quinolate was considered to be one of the most stable and highly fluorescent compounds in the class of metal chelates; (2) aluminum quinolate was known to form smooth thin films; and (3) an evaporated film of aluminum quinolate was microcrystalline with a grain size  $<500\text{\AA}$ . Accordingly, aluminum quinolate became the material of choice as the host material for electroluminescent layers, at least for generating red and green pixels. The extensive use of aluminum quinolate by OLED manufacturers is confirmed in a paper by Rajeswaran et al., *Journal of Chemical Crystallography*, Vol. 35, No. 1, January 2005 (a copy of which accompanies the Supplemental IDS). As stated in the Rajeswaran et al. publication:

“The interest in organic materials for use in organic light-emitting diodes (OLEDs) began with the pioneering report of efficient green electroluminescence from  $\text{Alq}_3$ , tris(8-hydroxyquinoline) aluminum by Tang and Van Slyke. After more than 17 years of intense research and development in OLEDs,  $\text{Alq}_3$  continues to be the most widely used electroluminescent material in OLED chemistry.  $\text{Alq}_3$  is used in electron transport and/or electron injecting layers in multiplayer device structures and also as an effective host material for various dyes.”

The heart of the technical problem to which the present application is directed is to find a material that is superior to aluminum quinolate for use as an electroluminescent host material. The difficulty in solving this problem is demonstrated by the long period of time

over which aluminum quinolate has remained the predominant material for such applications. In relation to a two-component electroluminescent layer, as now claimed, the newly-cited Cok '231 and Fukuyama '976 references give no guidance or suggestion whatsoever as to what might be a generally better host material than aluminum quinolate.

The present inventors have made the surprising and unobvious finding that organo metal complexes in which the metal has a valency greater than 3, such as zirconium quinolate and hafnium quinolate, have a number of properties that, judged in combination, make these materials that can be advantageously used in place of aluminum quinolate as the host material in a two-component electroluminescent layer of an OLED, resulting in surprisingly improved performance. These materials can also be used as an electron transport layer. The electroluminescent device performance of devices fabricated according to this invention has been found to be markedly superior to comparable devices using aluminum quinolate, as clearly shown by the application examples and by the comparative graphs of Figs. 37/49 to 47/49. No teaching in Cok '231 or Fukuyama '976 even suggests these unexpectedly superior results. In particular, in this invention:

(a) Zirconium and hafnium quinolates have been found in this application to serve as a like-for-like replacement for aluminum quinolate and thus can be substituted without requiring significant manufacturing process or equipment changes.

(b) Zirconium and hafnium quinolates have been found in this application to be useful with at least some of the same dopants as are used with aluminum quinolate. It has been found in this connection that the HOMO and LUMO levels for zirconium quinolate and aluminum quinolate are similar – the HOMO level for aluminum quinolate being  $-7.5\text{eV}$  and that for zirconium quinolate being  $-5.6\text{eV}$ , and the LUMO level of both compounds being

-2.eV. Cok '231 and Fukuyama '976 completely fail to recognize these important material characteristics of zirconium quinolate in these applications.

(c) Zirconium and hafnium quinolates have been found in this application to exhibit a higher electron mobility than aluminum quinolate, which in turn increases the current efficiency of an OLED fabricated according to the present invention. The reason for this increase is believed to be as follows: An electroluminescent device can be viewed as a diode which is forward-biased when the anode is at a higher potential than the cathode. Under these conditions, the anode injects holes (positive charge carriers) into the luminescent layer, while the cathode injects electrons into the luminescent layer. The injected holes and electrons each migrate toward the oppositely charged electrode, which results in hole-electron recombination within the luminescent layer. When a migrating electron drops from its conduction potential to a valence band in filling a hole, energy is released as light. Maximum light output for a given current (i.e., maximum current efficiency) requires a balance between holes and electrons. Typically in OLEDs the majority carriers in are holes, thereby limiting the luminescent response. By providing for an increase of electron mobility in the electroluminescent layer, however, the surprising result is an improved balance between holes and electrons and hence an unexpected improvement in luminous efficiency. These performance improvements are clearly seen in Table 1 of the subject application and in Figs. 37/49 to 47/49. In this respect as well, Cok '231 and Fukuyama '976 utterly fail to recognize these important material characteristics of zirconium quinolate and hafnium quinolate in these applications.

(d) It has been found in this application that there is a link between balanced majority and

minority carriers and device lifetime. Specifically, the change from aluminum quinolate to zirconium quinolate as host material for the electroluminescent layer in accordance with this invention has surprisingly been found to improve device lifetime. As shown by the data in this application, the fall-off of luminance over time occurs less quickly for devices fabricated according to this invention as compared to similar devices where the host is aluminum quinolate, and this is true regardless of whether the electron transport layer is zirconium quinolate or aluminum quinolate. This finding is particularly surprising in view of prior art teachings (e.g., in Fukuyama '976 – as discussed hereinafter) that suggest exactly the opposite result! Cok '231 and Fukuyama '976 thus completely fail to recognize or even suggest that a dramatic and unexpected improvement in luminescent life can be obtained with zirconium quinolate as the host material in accordance with this invention.

(e) Zirconium quinolate has no dipole moment which, as explained in earlier responses, gives rise to improved color characteristics. The improved CIE coordinates for doped aluminum quinolate reported in this application indicate a more saturated green. The emission of the doped zirconium quinolate device of this invention, by comparison, is much sharper and of higher intensity than for an aluminum quinolate-based device. Nowhere do Cok '231 or Fukuyama '976 disclose or even suggest the advantage of the significantly better color that is realized in the two-component zirconium quinolate devices according to this invention.

Use of zirconium or hafnium quinolate as the electroluminescent host therefore provides a solution to the long-standing problem of finding a better replacement for aluminum quinolate in such systems. Furthermore, the attributes of zirconium or hafnium quinolate which, in combination, make these materials especially suitable for use as a

replacement for aluminum quinolate host material, were completely unrecognized prior to the present invention and would never have been arrived at by a person skilled in the art working from the state of the art, specifically from the teachings of the Cok '231 or Fukuyama '976 references, for the reasons discussed hereinafter.

2. The Cok '231 Reference Does Not Teach or Suggest the Claimed Invention.

The Cok '231 reference does not teach or suggest the recited features or the recited device performance characteristics in the pending claims, nor would those features or characteristics be inherent in devices fabricated according to the teachings of Cok '231. In particular, Cok '231 is silent regarding the claimed performance characteristics of “wherein said device has the characteristics of a higher luminance efficiency measurable as  $\text{cd A}^{-1}$ , a greater luminance measurable as  $\text{cd m}^{-2}$  at  $20 \text{ mA cm}^{-2}$ , and a reduced turn-on voltage compared with a similar device in which said metal quinolate is aluminum quinolate.” None of these claimed features are taught by Cok '231, but the Examiner has apparently simply ignored these claim recitations.

The Cok '231 reference discloses organic electroluminescent devices comprising a light-emitting layer which “can be comprised of a single material, but more commonly consists of a host material doped with a guest compound or compounds....” (Cok '231 at col. 7, lines 63-67). A list of possible “host” materials is presented at col. 8, lines 30-49 of Cok '231, and a list of possible fluorescent dopants is presented at col. 8, lines 56-62.

In para. 10 of the Office Action, the Examiner referenced col. 8, lines 30-49 of Cok '231 to support his statement that this reference teaches “zirconium or hafnium quinolate as the host material....” This statement is clearly incorrect, however, because there is no

mention in Cok '231 of "hafnium quinolate" or any other hafnium compounds. Claims 65, 76 and 77 are now specifically directed to apparatus or methods reciting "hafnium quinolate" as the metal quinolate in the electroluminescent layer. These claims are certainly not anticipated by Cok '231.

As noted above, the present invention is based on the novel and surprising discovery that unexpectedly improved electroluminescent performance can be realized by using an organo metal complex of metals with a valency greater than 3 in the electroluminescent layer of an electroluminescent device. It is well recognized that the discovery that a certain, narrowly-defined set of materials (e.g., organo metal complexes of metals with a valency greater than 3) within a larger group (e.g., the set of all organo metal complexes) has unexpected and especially advantageous properties or uses as compared with the rest of the group constitutes a patentable invention.

Thus, in this case, at page 2, lines 9-10, the Specification of this application states: "We have now surprisingly found that the use of organo metal complexes of metals with a valency of greater than 3, e.g., of 4, 5 or higher, give improved performance...." At page 2, lines 24-25, the Specification further states: "Preferably the metal M is a transition metal such as titanium, zirconium or hafnium in the four valency state or vanadium, niobium or tantalum in the five valency state." Further, at page 22, lines 25-28, the Specification states: "The improved performance of the organo metallic quinolates where the metal has a valency greater than 3 compared with the state of the art, aluminum quinolate, is particularly shown in the [improved] efficiency of the electroluminescent [layers of this invention] although there is [also] an improvement in a range of [other] properties, e.g., lifetime, stability, etc."



Although the art in this field recognizes generally that a material such as zirconium quinolate can be used in the electroluminescent layer of an electroluminescent device, none of that art teaches or in any way suggests the unexpectedly improved performance that can be realized by: (1) choosing an organo metal complex wherein the metal has a valency greater than 3; (2) pairing that >3 valency organo metal complex with a fluorescent dopant selected from a limited group of dopant materials that cooperate synergistically with the organo metal complex to produce “optimal spectral coupling” (as discussed at page 4, line 4 to page 5, line 24 of the Specification); and (3) restricting the proportion of the selected dopant to a particular mole percent range. The Examiner is required to read the claims of this application consistently with the Specification, and the teachings of the references must be fairly and reasonably interpreted relative to the state of art at the time of the invention. As discussed in detail above in part (B)(1), at the time of this invention, aluminum quinolate was the preferred host material for EL device applications. Nothing in Cok ‘231 suggests anything different.

The pending claims are directed to a particular combination of zirconium or hafnium quinolate (as the >3 valency organo metal complex), a dopant selected from a narrowly defined group of fluorescent materials, and a particular mole% range of dopant to metal complex. This particular combination is neither disclosed nor suggested in any way by Cok ‘231.

It is true that Cok ‘231 identifies “metal complexes of 8-hydroxyquinoline” as constituting “one class of useful host compounds capable of supporting electroluminescence” (col. 8, lines 30-32), but no preference whatsoever is expressed for this group of materials compared with other types of host compounds. Among a list of nine examples of such “metal

complexes of 8-hydroxyquinoline,” only one is a zirconium compound (col. 8, lines 48-49) and none is a hafnium compound.

The zirconium compound is the only one of the nine Cok ‘231 examples in which “the metal has valency greater than 3”. Consistent with the state of the art in this field prior to this invention (preferring aluminum quinolate as the “host material”), three of the nine Cok ‘231 examples are varieties of aluminum quinolate. Besides aluminum, other valency = 3 metals in the Cok ‘231 list include indium and gallium. The Cok ‘231 list also includes metal quinolates where the metal has a valency less than 3 – such as magnesium (valency = 2), and zinc (valency = 2) – and even lithium (valency = 1), but there are no others with a valency >3.

Cok ‘231 indicates no preference whatsoever among these nine examples of quinolate host compounds, and the reference teaches no reason why a person of ordinary skill in this art would choose one of these materials instead of another. Certainly there is no teaching in Cok ‘231 that the single >3 valency metal quinolate listed (the zirconium compound) would be associated with improved electroluminescent performance.

Similarly, there is no reason based on the teachings of Cok ‘231 for a person to choose certain of the many fluorescent dopant compounds listed at col. 8, lines 56-62 over others in the list. The independent claims now pending recite specific classes of dopant materials that are to be paired with the zirconium or hafnium quinolate in the electroluminescent layer. Claims 77, 88, 91 and 92 now specifically recite diphenylquinacridine (DPQA) as the dopant, which is a dopant that is not taught by Cok ‘231. ~~These~~ claims therefore cannot be considered to be ~~anticipated~~ by the reference.

For all of the foregoing reasons, the rejection of claims under 35 U.S.C. §102(e) based on Cok '231 should be reconsidered and withdrawn.

**C. Sec. 102(b) Rejection – Fukuyama '976**

Claims 64-67, 69, 71-79, 81 and 83-90 were rejected under 35 U.S.C. §102(b) as being anticipated by the Fukuyama '976 Japanese patent publication (para. 11 of the Office Action of March 12, 2010). Reconsideration of this rejection is respectfully requested for the following reasons:

As thoroughly discussed in part (B)(1) above, at the time of this invention aluminum quinolate was the standard and preferred host material for use in the electroluminescent layers of electroluminescent devices. Other materials, including zirconium quinolate, were known to function in such applications, but because such other materials were either believed to have disadvantages, or at least to offer no particular advantages, there was typically no reason for one of ordinary skill in this art to choose anything other than aluminum quinolate. The teachings of Fukuyama '976 completely reinforce this description of the state of the art at the time of this invention.

The Fukuyama '976 reference discloses electroluminescent devices comprising an electron transport layer and/or a luminous layer including a substituted or unsubstituted metal quinolate which, in at least some examples, can be zirconium quinolate. In Example 1 (at para. [0027] of the reference), aluminum quinolate ( $\text{Alq}_3$ ) was used as the “luminous layer 4” and a zirconium compound was used only as the “electron transport layer 5”. In Example 2 (at para. [0029] of the reference), aluminum quinolate doped with a coumarin dopant was used as the “luminous layer 4”.

In Example 3 (at para. [0032] of the reference), a zinc (valence = 2) compound was used as the “luminous layer 4”. In Example 5 (at para. [0033] of the reference), a zirconium compound is used as a part of the “luminous layer 4,” but it is used together with a “JISUCHIRIRU arylene derivative (DPVBi)” material as a dopant. Example 5 of Fukuyama ‘976 is the only example in which the host material comprises a zirconium compound, and it is also the only example in which an organo metal complex where “the metal has a valence greater than 3” is used in “luminous layer 4”.

There is no mention whatsoever in Fukuyama ‘976 of hafnium quinolate or any other hafnium compounds. Claims 65, 76 and 77 are now specifically directed to apparatus or methods reciting “hafnium quinolate” as the metal quinolate in the electroluminescent layer. These claims are certainly not anticipated by Fukuyama ‘976.

As noted above, Fukuyama ‘976 only teaches the combination of zirconium quinolate with the arylene derivative DPVBi as dopant for the luminous layer 4. But, arylene derivatives, and specifically DPVBi, are outside the scope of the specific classes of dopant materials that are now recited in the independent claims. Claims 77, 88, 91 and 92 now specifically recite diphenylquinacridine (DPQA) as the dopant, which is certainly not taught by Fukuyama ‘976. These claims therefore cannot be considered to be anticipated by the reference.

Furthermore, the Fukuyama ‘976 reference can be interpreted as clearly teaching away from Example 5 and the use of a zirconium compound as the host material. Table 1 of Fukuyama ‘976 compares key performance characteristics of electroluminescent devices fabricated in accordance with Examples 1 to 6 of the reference. Table 1 clearly shows that the performance of the device of Example 5 (using the zirconium compound as host material)

is inferior in various respects to the devices of other Examples (using host materials where the metal has a valence of 3 or less), and specifically inferior to the devices using aluminum quinolate as the host material – e.g., Examples 1 and 2.

Thus, Table 1 of Fukuyama '976 shows that the device of Example 5 has a higher driving voltage (6.3V) compared with the devices of every other Example in the Table. Lower driving voltage is a preferred performance characteristic for these devices. Table 1 also shows that the device of Example 5 has a dramatically shorter half life (only 30 hours) compared with the devices of every other Example in the Table. The device of Example 2, for instance, using aluminum quinolate and a dopant, demonstrates a half life of 1500 hours! Longer life is a preferred performance characteristic for these devices.

Based on the results shown in Table 1 of Fukuyama '976, no one of ordinary skill in this art would ever be motivated to use zirconium quinolate in place of aluminum quinolate as the host material.

For all of these reasons, the rejection of claims under 35 U.S.C. §102(e) based on Fukuyama '976 should be reconsidered and withdrawn.

**D. Sec. 103(a) Rejection – Cok '231**

Claims 70-72, 82-84 and 90 were rejected under 35 U.S.C. §103(a) as unpatentable over Cok '231 (para. 14 of the Office Action).

Applicants repeat and incorporate herein the arguments set forth in parts (B)(1) and (B)(2) above with respect to the Cok '231 reference. For the same reasons argued above, Cok '231 does not teach or in any way suggest the claimed device or method wherein the

electroluminescent layer consists of a specific combination of zirconium or hafnium quinolate and a dopant selected from the recited species.

Indeed, as argued above in part (C), Table 1 of the Fukuyama '978 reference would lead one of ordinary skill in this art away from selecting a zirconium host material and back to the tried and true standard – aluminum quinolate. All of the Examiner's assertions in para. 14 of the Office Action regarding "obviousness" are now clearly seen as being based on improper "hindsight" reconstruction because nothing in the art would motivate someone to make the selections and substitutions that the Examiner asserts "would be obvious".

For all of these reasons, the rejection of claims under 35 U.S.C. §103(a) based on Cok '231 should be reconsidered and withdrawn.

**E. Sec. 103(a) Rejection – Fukuyama '976 / Kido '209**

Claims 68, 70, 80 and 82 were rejected under 35 U.S.C. §103(a) as unpatentable over Fukuyama '976 in view of Kido '209 (para. 15 of the Office Action).

Applicants repeat and incorporate herein the arguments set forth in part (C) above with respect to the Fukuyama '976 reference. For the same reasons argued above, Fukuyama '976 does not teach or in any way suggest the claimed device or method wherein the electroluminescent layer consists of a specific combination of zirconium or hafnium quinolate and a dopant selected from the recited species. Kido '209 appears to be cited here for its teaching of  $\alpha$ -NPB as a hole transport material in electroluminescent devices and for its teaching of lithium quinolate in an electron injection layer. However, Kido '209 does not make up for the fundamental deficiencies in the teachings of Fukuyama '976 relative to the pending claims. Specifically as argued above, in part (C), Table 1 of the Fukuyama '978

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reference would lead one of ordinary skill in this art away from selecting a zirconium host material and back to the tried and true standard – aluminum quinolate.

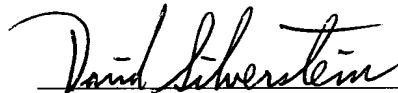
For all of these reasons, the rejection of claims under 35 U.S.C. §103(a) based on Fukuyama '976 in view of Kido '209 should be reconsidered and withdrawn.

#### **SUMMARY AND CONCLUSIONS**

For all of the foregoing reasons, Claims 64-92 now pending should be considered in condition for allowance and an early notice thereof is earnestly requested.

Respectfully submitted,

Date: July 10, 2010  
Andover-IP-Law  
44 Park Street, Suite 300  
Andover, MA 01810  
Telephone: (978) 470-0990  
Facsimile: (978) 470-0993

  
\_\_\_\_\_  
David Silverstein  
Registration Number 26,336  
Attorney for Applicants